

IN THE CLAIMS

Please amend the claims as indicated below, deleting text marked with strike through and adding the underline text. This set of claims replaces all previous claim sets.

1. (Currently Amended) Organic solutions containing metal(IV) salts and oxoacids of phosphorus from which, after evaporation of ~~[[the]]~~ a solvent, insoluble compounds of general composition $M(IV)(O_3P-G)_{2-n}(O_3P--R^1--X)_n$ can be obtained, where M(IV) is a tetravalent metal, -G is a generic inorganic or organic group, --R¹-- is an organic group, --X is an acid group and n is a coefficient ranging from 0 to 1.5.

2. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the ~~anion of the~~ tetravalent metal salt is ~~preferably chosen among~~ an anion and is selected from the group consisting of carboxylates, chlorides and alkoxides.

3. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the tetravalent metal salt is ~~preferably chosen between~~ selected from the group consisting of Zr, Ti, Sn and Ce or their mixture.

4. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the tetravalent salt is ~~preferably the~~ zirconyl propionate or chloride.

5. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the group -G is ~~preferably chosen among the~~ selected from the group consisting of acid groups --OH; --R²--SO₃H and --R²--PO₃H₂[[,]]; and where --R²-- is an organic group with ~~preferably linear chain such as~~ selected from the group consisting of --(CH₂)_m-- and --(CF₂)_m--.

6. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the group --R¹-- is an ~~arylen~~ arylene group ~~chosen preferably among~~ selected from the group consisting of --C₆H₄--; --C₆H₄--CH₂-- and --C₆H₄--CF₂--.

7. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the acid group --X is ~~chosen between~~ selected from the group consisting of --SO₃H, --PO₃H₂ and --COOH.

8. (Currently Amended) The organic ~~Organic~~ solutions of claim 1 wherein the organic solvent is ~~chosen among the a~~ protonable solvent or solvents selected from the group consisting of , especially N,N-dimethylformamide, N-methyl-2-pyrrolidone, dioxane, dimethylsulfoxide, acetamide, acetonitrile, various alkanols and mixtures thereof and/or ~~their mixtures, commonly used for dissolving the proton~~ conducting ionomers of the state of art.

9. (Currently Amended) ~~Use of the organic solutions of any of the preceding claims~~ The process for the insertion of nano-particles of tetravalent metal salts, ~~preferably phosphate-phosphonates,~~ within the pores of polymeric or inorganic porous membranes comprising:

a) preparing the organic solution of claim 1 which, at the same time, may also contain a polymer and/or an ionomer; b) impregnating the porous membrane with the solution; c) eliminating the solvent; d) repeating the steps b) and c) until a partial or complete pore filling is obtained; and

wherein the tetravalent metal salts are phosphate-phosphonates.

10. (Cancelled)

11. (Currently Amended) A method of preparing proton ~~Proton~~ conducting composite membranes ~~or mixtures of said compounds with a proton conducting ionomer and especially prepared by using the solutions of claims 1-8,~~ comprising:

a) preparing the organic solution of claim 1 which, at the same time, may also contain a polymer and/or a proton conducting ionomer; b) impregnating a polymeric or inorganic porous membrane with the solution; c) eliminating the solvent; d) repeating steps b) and c) until a partial or complete pore filling is obtained; and

wherein the tetravalent metal salts are phosphate-phosphonates.

12. (Currently Amended) The proton ~~Preton~~ conducting composite membranes of claim 11 wherein the polymeric porous membrane is a polymer ~~preferably chosen between these~~ made of chemically and/or thermally stable polymers, especially selected from the group consisting of, polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polyesters, polyethersulfones and fluoroelastomers.

13. (Currently Amended) The proton ~~Preton~~ conducting composite membranes of claim 11 wherein the pore dimensions of the porous membranes are ~~preferably~~ in the range 0.02-20 μm , ~~especially 0.1-10 .mu.m, preferably 0.4-2 .mu.m and~~ [[the]] a porosity >10%, especially >50%, preferably 65- 90%.

14. (Currently Amended) The proton ~~Preton~~ conducting composite membranes of claim 11 wherein the phosphate-phosphonates, ~~for the filling of pores are chosen between~~ selected from the group consisting of $\text{Zr}(\text{O}_3\text{P}-\text{CH}_2-\text{PO}_3\text{H}_2)_2$ and compounds of the series $\text{Zr}(\text{O}_3\text{P}-\text{OH})_{2-n}(\text{O}_3\text{P}-\text{C}_6\text{H}_4-\text{SO}_3\text{H})_n$, and $\text{Zr}(\text{O}_3\text{P}-\text{C}_6\text{H}_4-\text{SO}_3\text{H})_{2-n}(\text{O}_3\text{P}-\text{CH}_2-\text{PO}_3\text{H}_2)_n$, with n in the range 0.1-1.5.

15. (Currently Amended) A method of preparing conducting composite ~~Composite~~ membranes made up of a porous ceramic membrane, comprising:

a) preparing the organic solution of claim 1; b) impregnation of a porous ceramic membrane with the solution; c) eliminating the solvent; d) repeating steps b) and c) until a partial pore filling is obtained;

wherein the pores are partially filled with a tetravalent metal salt; and

wherein the membranes exhibit, ~~preferably phosphate-phosphonate, according to claim 1 exhibiting catalytic activity.~~

16. (Currently Amended) The composite ~~Composite~~ membranes of claim 15 wherein the tetravalent metal salts, ~~preferably is a phosphate-phosphonate selected from the group consisting of~~ $\text{Zr}(\text{O}_3\text{P}-\text{CH}_2-\text{PO}_3\text{H}_2)_2$ and compounds of the series $\text{Zr}(\text{O}_3\text{P}-\text{OH})_{2-n}(\text{O}_3\text{P}-\text{C}_6\text{H}_4-\text{SO}_3\text{H})_n$, and $\text{Zr}(\text{O}_3\text{P}-\text{C}_6\text{H}_4-\text{SO}_3\text{H})_{2-n}(\text{O}_3\text{P}-\text{CH}_2-\text{PO}_3\text{H}_2)_n$, with n in the range 0.1-1.5, ~~with catalytic activity is chosen among those reported in claim 14.~~

17. (Currently Amended) ~~Use of the organic solutions of any of claims 1-8 for~~ A method for the preparation of nano-polymers constituted by nano-particles of tetravalent metal salts, comprising,

a)preparing the organic solution of claim 1 which, at the same time, may also contain an organic or organic polymer or polymers and thereby producing a preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers soluble in the same solvents, and b) elimination of the solvent ; and

wherein the tetravalent metal salts are phosphate-phosphonates.

18. (Currently Amended) ~~Use of the organic solutions of any of claims 1-8 for the preparation of nano-polymers~~ The method of claim 17 wherein the organic polymeric matrix is that of a proton conducting ionomer.

19. (Cancelled)

20. (Currently Amended) The method for preparing the ~~preparation of~~ nano-polymers and nano-ionomers of claim ~~[[19]]~~ 18 wherein the elimination of the solvent is preferably performed by evaporation or with a non-solvent of the polymer or ionomer.

21. (Currently Amended) Nano-polymers produced by the method of claim 17~~constituted by nano particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers.~~

22. (Currently Amended) ~~Nano-polymers~~ The nano-polymers of claim 21 wherein the ~~polymeric~~ matrix is ~~that of~~ a synthetic ionomer selected from the group consisting of the state of the art preferably chosen among perfluorosulphonic polymers, sulfonated polyetherketones (sPEK), sulfonated polyethersulfones and sulfonated polyvinylidenefluoride (sPVDF).

23. (Currently Amended) ~~Nano-polymers~~ The nano-polymers of claim 21 wherein the nano-particles of tetravalent metal salts, ~~preferably phosphate phosphonates,~~ dispersed in the polymeric matrix ~~are chosen among those exhibiting~~ exhibit proton conductivity $>10^{-2}$ S cm⁻¹ at 70°C. and 95% relative humidity.

24. (Currently Amended) ~~Nano-polymers~~ The nano-polymers of claim 21 wherein the nano-particles of tetravalent metal salts, ~~preferably phosphate phosphonates,~~ are ~~those constituted by the same compounds of claim 14~~ selected from the group consisting of $Zr(O_3P--CH_2--PO_3H_2)_2$ and compounds of the series $Zr(O_3P--OH)_{2-n}(O_3P--C_6H_4--SO_3H)_n$, and $Zr(O_3P--C_6H_4--SO_3H)_{2-n}(O_3P--CH_2--PO_3H_2)_n$, with n in the range 0.1-1.5.

25. (~~Currently Amended~~) ~~Use of the organic solutions of claim 1 for~~ A method for the preparation of membranes constituted by nano-polymers of claim 21, comprising:

a)preparing the organic solution of claim 1 which, at the same time, may also contain an organic or organic polymer or polymers and thereby producing a matrix of organic or inorganic polymers soluble in the same solvents, and b) eliminating of the solvent ; and

wherein the tetravalent metal salts are phosphate-phosphonates.

26. (~~Currently Amended~~) ~~Nano-ionomeric proton conducting membranes constituted by nanopolymers~~ The method of claim 21, wherein the membranes are nano-ionomeric proton conducting membranes.

27. (~~Currently Amended~~) ~~Use of the organic solutions of claim 1~~ A method for an easy insertion of a large variety of lamellar nano-particles of tetravalent metal salts, preferably phosphate phosphonates, in the membrane/electrode interfaces of PEM FCs, comprising preparing the organic solutions according to claim 1 and eliminating the solvent.

28. (Currently Amended) ~~Use of the organic solutions of claim 1 with~~ A method according to claim 27, further comprising the addition of ionomers and/or other proton conducting compounds soluble in the same solvents, ~~for an easy insertion of a large variety of lamellar nanoparticles of tetravalent metal salts, preferably phosphate phosphonates, in mixture with other proton conducting compounds in the membrane/electrode interfaces of PEM FCs.~~

29. (Currently Amended) ~~Use of proton conducting membranes constituted by inorganic or polymeric porous membranes with pores filled with tetravalent metal salts, preferably phosphate phosphonates, of any of claims 11-16 and of membranes constituted by~~ An electrochemical device comprising the nano-polymers of claim 26, ~~in electrochemical devices.~~

30. (Currently Amended) ~~Use of proton conducting membranes of claim 29 in~~ The electrochemical devices specifically planned device of claim 29, wherein said device is designed for generating electrical energy from the oxidation of a fuel.

31. (Currently Amended) ~~Use of proton conducting membranes of claim 29 in~~ A fuel cell cells specifically planned for electrical vehicles and/or for portable electrical devices comprising the electrochemical device of claim 29.

32. (Currently Amended) ~~Use of composite membranes of claim 29 for improving the global performance of ionomeric membranes of the state of the art in hydrogen, indirect~~ Indirect methanol and direct methanol fuel cells comprising the electrochemical device of claim 29.

33. ~~Use of membranes of claim 11 in A~~ catalytic membrane reactors comprising the proton conducting composite membranes produced according to claim 11.